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3D printing with light: Towards additive manufacturing of soft, electroactive structures

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ABSTRACT

Soft electroactive devices such as dielectric elastomer actuators have been the subject of research for several decades. One of the reasons they have not found many industrial applications to date is the challenge of manufacturing such devices cost-effectively. 3D printing is now widely used to cut manufacturing time and cost in many industries, but functional and soft materials for 3D printing are still very limited. Here we present a process that could be used to 3D print functional soft, electroactive devices like dielectric elastomer actuators and sensors. We propose a simple, low-cost 3D printing platform that uses direct ink writing of elastomer composites with low filler loading of carbon-based nanoparticles. These composites allow the deposition of smooth, uniform layers as well as rapid curing of printed materials with UV light. A diode laser is then used to induce a chemical transformation of the elastomer to create conductive patterns for electrodes with arbitrary shapes and high detail. The process is still limited by the low elasticity of the laser induced electrode material but if more suitable materials can be found, this process could dramatically reduce the time, cost and complexity involved in manufacturing dielectric elastomer devices while at the same time greatly increasing the possible geometric and functional complexity of the 3D printed devices.

Keywords: Dielectric elastomers, silicone rubber, elastomer composites, direct ink writing, UV curing, laser scribing, graphene oxide, graphite nanoparticles

1. INTRODUCTION

Soft, electroactive structures have great potential for many applications in fields such as robotics, wearables, haptic interfaces and medical devices, wherever structural compliance and shape-changing capability is desirable. Dielectric elastomers (DEs) in particular show great promise in all of these areas. Their actuation, sensing and energy harvesting capabilities make them highly versatile, soft electromechanical transducers.[1] Despite many advances in recent years, the major limiting factor for the wide adoption of DE technology still lies in suitable soft materials and manufacturing processes.[2]

DE devices generally require the deposition of thin, defect-free layers of alternating dielectric and conductive materials. Many applications require large numbers of parallel layers, increasing the risk of failure due to defects introduced during manufacturing. There is also only a limited set of known, suitable materials for both the soft dielectric and compliant electrodes. Most often, silicone elastomer is used for its low elastic modulus, low viscous loss and good dielectric breakdowns strength.[1] Electrode materials range from conductive carbon or silver particles over vapor-deposited thin metal films to ionic gels. Most commonly used is some form of conductive carbon such as carbon black, graphite, graphene or carbon nanotubes, either on their own, as a paste or incorporated in an elastomer matrix.[3] Lotz et al. were the first to demonstrate fully automated manufacturing of stacked DE actuators by repeated spin coating of liquid silicone

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rubber and spray deposition of ink containing graphite particles.[4] However, this process still requires pre-fabricated, part-specific masks for electrode deposition and cannot control the shape of the elastomer layers.

A 3D printing process capable of manufacturing DE devices would be highly desirable as it can produce arbitrary shapes and configurations of DEs without any need for molds or part-specific tools and is also fully automated. 3D printing technology has been advancing rapidly over the past decade and new processes and materials are being developed to 3D print functional structures[5] but particularly printing with soft, functional materials with high enough reliability for DE fabrication remains challenging. In this work we propose a low-cost platform that combines 3D printing with a UV-curable elastomer with laser scribing to produce conductive patterns on printed layers, with the potential to produce fully functional DE actuators and sensors in a single automated procedure.

2. RESULTS AND DISCUSSION

To 3D print soft electroactive devices, we apply a simple direct ink writing technique. In this process, a continuous bead of material in liquid or paste form is deposited from a moving nozzle to "draw" each layer of the printed part. The 3D printer used in our research is a custom-built model based on the open-source Prusa i3 design. The printer is equipped with two extruders suitable for dispensing high-viscosity fluids in a range between 50 Pa·s and 500 Pa·s. This setup allows alternating deposition of two different materials e.g., for dielectric and electrodes of a DE device. Images of the printer and a detailed view of the print head is shown in Figure 1. The printhead has lifting nozzles to avoid contamination of a layer with material from the unused extruder which could immediately cause device failure in case of the dielectric getting contaminated with conductive electrode material. The printer uses tapered nozzles with 0.41 mm diameter which are suitable for printing layers from 100 µm to 300 µm thickness. The test prints shown in this work have been printed with 200 µm layer thickness but printing at 100 µm layer height has been demonstrated successfully. Printing even thinner layers should be possible but will require a smaller nozzle to maintain constant flow at low extrusion rates.

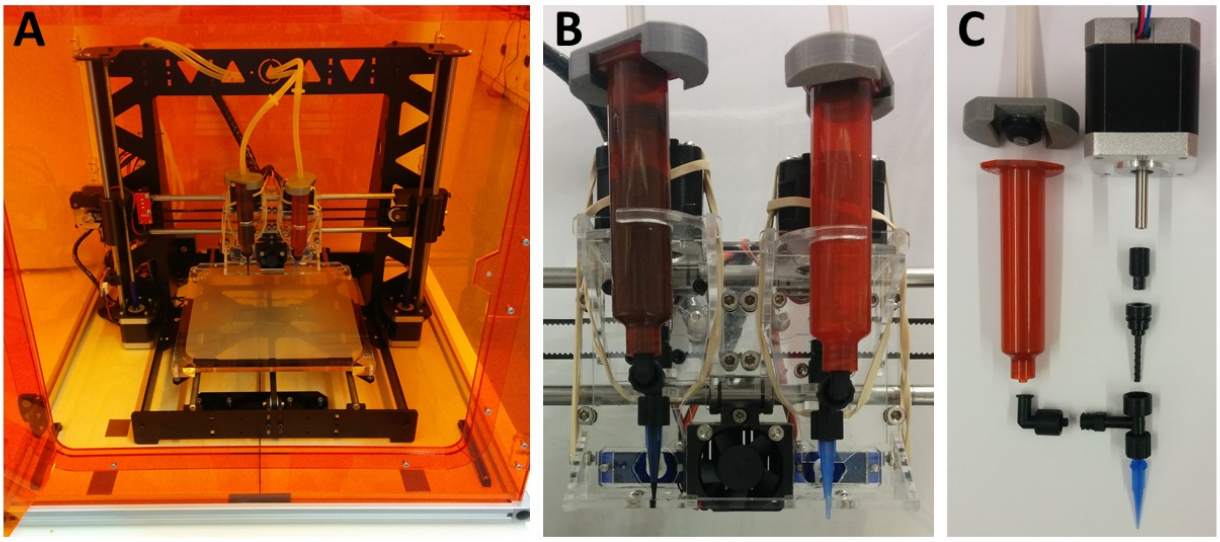

Figure 1. The 3D printer used in this work (A), inside its UV safety enclosure. The print head (B) uses two disposable auger valves (C) to dispense liquid silicone rubber or other viscous materials such as carbon grease. Material is held in 10 mL disposable syringes. The syringes are kept under constant overpressure between 100 mbar and 500 mbar to aid material flow to the auger valve.

Attempts were made to print dielectric elastomer devices using conductive carbon grease as electrode material. Carbon grease is commonly used in the manual manufacture of DEAs as compliant electrode material. Figure 2C shows a printed test piece with a single layer of carbon grease encased in clear silicone rubber. However, because the carbon grease remains liquid after deposition, silicone deposited on top of the carbon grease tends to displace the grease which

then protrudes between adjacent lines of silicone or is picked up by the nozzle and smeared along the path of the print head, as is visible in the bottom half of Figure 2C. This leads to non-uniform layers of silicone and insufficient insulation between conducting layers of a printed dielectric elastomer device, making carbon grease an unsuitable material for this printing process.

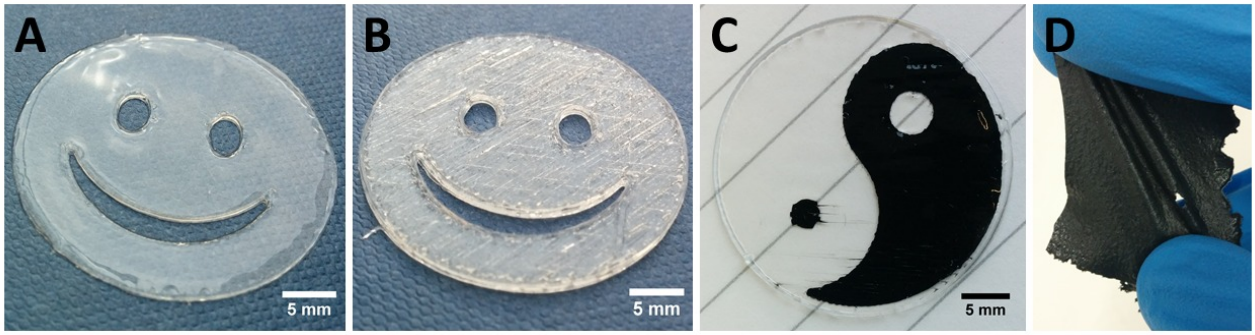

Figure 2. Test prints demonstrating the effect of settling: letting the uncured silicone material flow for several seconds before curing results in a smooth, even surface (A). By comparison, the same shape printed with high-viscosity silicone that does not flow shows visible lines from the deposition (B). Two-material prints can be made with conductive carbon grease encased in layers of silicone (C), however, uncured conductive material can easily contaminate insulating layers during printing. Silicone rubber composites with graphite nanoparticles (D) are highly absorbent and not UV-curable, making them unsuitable for 3D printing.

As an alternative, UV-curable silicone filled with graphite nanoparticles (GNP) was prepared to test its suitability for 3D printing. Two samples GNP-10 and GNP-20, with 10 wt% and 20 wt% filler loading respectively, were prepared. Their properties are shown in table 1. Only sample GNP-20 was found to be electrically conductive. However, the high filler loading drastically increased the viscosity of the material and made it unsuitable for extrusion in our 3D printing platform. More importantly, the graphitic filler is highly absorbent in both the visible and UV spectrum and inhibits UV curing of the composite material. Even thin films (200 µm thickness) prepared from both samples showed no curing after exposure to UV light for as long as 24 hours. The composites can be heat cured instead but this process is much slower than UV curing and not feasible in a 3D printing process. Figure 2D shows that a (heat-cured) film of GNP-10 is fully opaque, making the composite unsuitable for UV curing.

Since most of the commonly used conductive fillers such as carbon black, graphite/graphene or carbon nanotubes are all highly absorbent, no composites made with such fillers will be UV-curable. (Silver nanowires are a possible candidate for making soft, transparent electrode materials[6,7] that might be UV curable but no such formulation was attempted, mainly due to the high cost of silver nanowires.) We therefore investigated an alternative approach that does not require deposition of an electrode material at all. By in-situ, laser-induced chemical transformation of the base silicone rubber, a layer of conductive material can be produced. It has been observed previously, that laser irradiation of silicone rubber can lead to combustion of the elastomer, producing domains of carbon and silicon compounds in the irradiated area.[8] Graphitic nanocrystals and multi-walled carbon nanotubes have been identified as products of laser-induced combustion and the composition of these products is dependent on laser intensity and other process parameters.[9,10] Our own experiments show that the residue from laser-induced combustion of silicone rubber is indeed electrically conductive. We therefore propose the use of laser scribing as a method to produce electrically conductive patterns on the surface of deposited silicone elastomer during the 3D printing process via chemical conversion of the silicone material.

It has been demonstrated that laser-induced combustion can be achieved by irradiation with a low-cost diode laser from a CD/DVD unit (785 nm).[11] In the works by Hautefeuille et al[9–11], carbon particles were applied to the surface or incorporated in the silicone elastomer to absorb the laser light and locally heat the elastomer to cause combustion. We found that laser induced combustion of the UV-curable silicone used in our 3D printing process is possible without incorporation of carbon particles using a 250 mW, 405 nm violet diode laser. However, initiation of the combustion was very unreliable on clear silicone and probably required some dust particle or other impurity on the surface to generate enough heat to initiate combustion. Once initiated, combustion continued consistently with laser irradiation, presumably due to the increased light absorption by the combustion products themselves.

To improve the reliability of the process, small amounts of GNPs were incorporated in the silicone elastomer to improve absorption of the laser. A silicone/graphene oxide (GO) composite material was also prepared. GO was chosen as a potentially interesting filler material because it can itself be converted to reduced graphene oxide (rGO) by laser irradiation under very similar conditions to those used for the combustion of silicone.[12] GO is generally less absorbent in the visible and near-UV spectrum than other carbon materials at the same concentration, potentially interfering less with UV curing. GO is also non-conductive and might therefore have a smaller impact on the dielectric breakdown strength of the composite. Laser reduction of GO increases both its absorbance and its electrical conductivity. We therefore hypothesized that GO, incorporated in the silicone rubber, might get reduced by laser irradiation and increase its absorption enough to initiate combustion at lower filler ratio than GNPs while at the same time increasing the conductivity of the combustion residue due to the presence of rGO.

Silicone/GO composites with 0.05 wt% (GO-005) and 0.1 wt% (GO-01) were prepared with UV-curable silicone. Two other composites with graphite nanoparticles at 0.03 wt% (GNP-003) and 0.6 wt% (GNP-006) mass fraction were also prepared for comparison. The composite samples and their properties are listed in Table 1. For GO-005, the filler did not have a notable effect on UV curing. A 200 µm film of the composite could be fully cured under the same conditions as pristine silicone (5 s/cm² UV exposure at 3.8 W). In contrast, for GO-01 UV curing was slowed down significantly. Even after repeating exposure six times (total exposure 30 s/cm²), a 200 µm film of the composite remained tacky and easily tore while peeling off the PET substrate (fully cured films peel off easily without tearing). GNP-003 also fully cured after normal exposure. GNP-006 could be UV cured but remained tacky after the first exposure, indicating that slightly longer exposure is required. After a second exposure, the composite was fully cured.

Table 1. Properties of silicone composite samples with varying ratios of GO and GNP fillers.

Sample	Filler material	Filler mass fraction	UV-curable	Bulk conductivity before laser scribing (S/m)	Sheet resistance after laser-scribing at 300 mm/min (kΩ□)
GO-005	GO	0.05 wt%	yes	$> 10^{-9}$ (not measurable)	100
GO-01	GO	0.1 wt%	no	$> 10^{-9}$ (not measurable)	-
GNP-003	GNP	0.03 wt%	yes	$> 10^{-9}$ (not measurable)	30
GNP-006	GNP	0.06 wt%	yes (~2x slower)	$> 10^{-9}$ (not measurable)	50
GNP-03	GNP	0.3 wt%	no	$> 10^{-9}$ (not measurable)	-
GNP-10	GNP	10 wt%	no	$7*10^{-6}$	-
GNP-20	GNP	20 wt%	no	30	-

The width, depth and composition of the combustion residue has been found to vary with laser power density and exposure time.[10] In our experiments, laser power was kept at maximum (250 mW) and only laser speed was varied. We found that laser scribing of all three UV-curable composites was more reliable and started occurring at higher scan speed of the laser, around 600 mm/min. On clear silicone, combustion was only observed at scan speed of 50 mm/min. We also observed a strong dependence of the properties of the laser-scribed layers on laser speed. Figure 3B shows patches of laser-scribed silicone at five different speeds between 500 mm/min and 100 mm/min on the three composites. At high speeds, combustion is intermittent and individual lines are visible whereas at lower speed, laser-scribed lines overlap and form a connected surface. Figure 3B also shows that the effect of laser scribing was weaker in GO-005 than in GNP-006 at the same speed, disproving the hypothesis that GO composites require a lower filler loading to achieve the same level of absorption.

The electrical resistance of the laser-scribed material also varied with laser speed. A plot of the sheet resistances measured for the laser-scribed patches visible in Figure 3B is shown in Figure 3C. The lowest sheet resistances were measured at 300 mm/min for all material samples and the values are listed in Table 1. Resistance consistently increased again for lower speeds, presumably due to a change in the composition of the combustion residue at higher temperatures.

Depth of the laser-scribed layers at 300 mm/min is around 10 µm as determined by focal length under a light microscope.

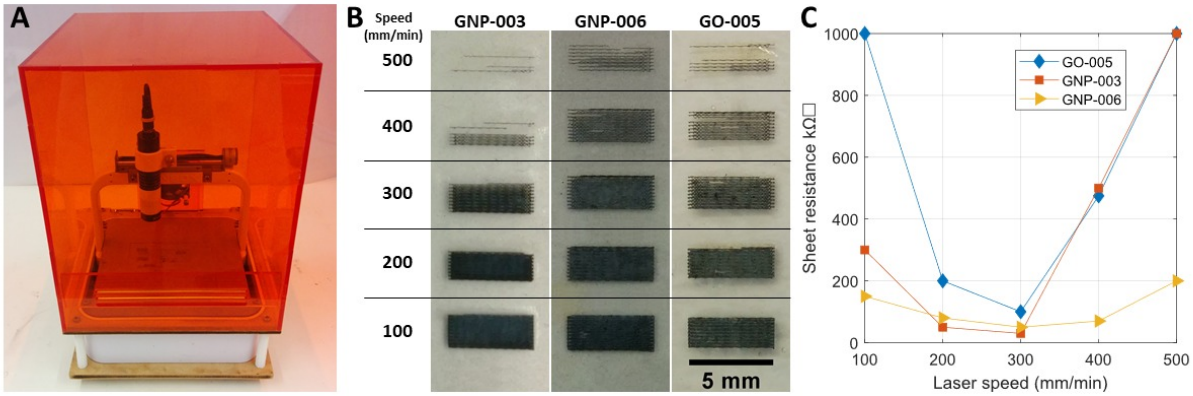

Figure 3. The laser engraver (A) used for laser scribing silicone rubber with varying fillers to produce conductive patterns (B). The width and composition of the laser-scribed silicone depends on the laser power and scanning speed. All patches were laser scribed with maximum power (250 mW) and varying speed between 100 and 500 mm/min. The graph (C) shows the corresponding sheet resistance measured for each of the laser-scribed patches in (B).

While the sheet resistance of laser-scribed silicone is in a similar range as other electrode materials for DEs[3] at rest, it was found that even very low strain or deformation led to a sharp increase in resistance. The resistance of a 10 x 10 mm patch of laser-scribed silicone (made from GNP-006) on a PET substrate was measured to be 1 MΩ at rest. The resistance increased to over 211 GΩ (the instrument limit) at the slightest convex bending (~5°, equivalent to roughly 0.2 % strain). When returned to rest after bending to 90°, the resistance reduced again to 40 MΩ. The resting resistance remained at this value after several more bending cycles. Concave bending (resulting in compression of the film) to 90° reduced the resistance to 2 MΩ. These observations indicate fracture of the conductive film at very low strain leading to separation of conductive domains and a loss of electrical conductivity. Relaxation or compression partially restore the interconnections between separated domains, but the film remains fractured, leading to a permanent increase in resistivity after the first bending cycle.

This behavior is not surprising, considering that combustion of silicone elastomer produces silicon dioxide, silicon carbide and silicon oxycarbide[11], all of which are very hard, brittle ceramic materials. Unfortunately, this makes laser-scribed silicone rubber unsuitable as electrodes for elastic actuators and sensors. It will be difficult to avoid the formation of these brittle silicon compounds while using silicone rubber as a base material, however, the laser-scribing process might be applicable to other kinds of rubber that form a higher fraction of conductive carbon species that also have higher elasticity/plasticity. For instance, the formation of highly conductive graphene layers by laser scribing polyimide films has already been documented.[13] Possibly, this property also extends to other elastomeric materials such as, for example, polyurethane. If this is the case, laser-scribing could become a viable process to produce conductive patterns on 3D-printed elastomer films for the manufacture of soft, electroactive devices.

Although laser scribing of silicone rubber does not produce viable electrodes for DE devices, we demonstrated the feasibility of laser scribing to produce thin conductive patterns on the surface of a 3D-printed elastomer. If more suitable materials can be found, integration of laser scribing into the 3D printing process, might allow the additive manufacturing of functional DE devices with arbitrary geometry and high detail in an extremely simple, low-cost process. Using this technique requires deposition of only one material, which drastically reduces the complexity of the 3D printer. The only tools required are one extruder capable of accurately dispensing medium- to high-viscosity fluids, a UV LED for curing, and a low-power diode laser for laser scribing. All the components for such a system are cheap and readily available. The entire system can be built for under $1000. The process requires no molds or masks and the material used can be free of organic solvents.

Potentially, the laser-scribing technique could even be used as an addition to a stereolithography (SLA) printing process, where a laser is used to successively cure very thin layers of liquid resin either at the surface or at the bottom of a reservoir to form a 3D object. Another laser, or the same laser at increased intensity, might then be used in a second step to laser-scribe the surface of the material to produce conductive layers. SLA printers have very high resolution but are generally limited to a single material. Laser scribing silicone might be a possible route to producing two-material functional parts on an SLA printer.

3. CONCLUSIONS

In this work, we have outlined a possible 3D printing process to manufacture dielectric elastomer (DE) devices using a very simple and low-cost direct ink writing technique. The process requires only a single material to be deposited and it uses two separate sources of light to cure the material and then induce chemical transformation to produce conductive electrode layers. We have demonstrated 3D printing of soft structures using UV-curable silicone rubber in a simple, low-cost direct ink writing process with layer thickness down to 100 μm and short curing times on the order of seconds per layer. By giving the deposited material time to flow and settle after deposition, very uniform layers can be achieved which are important for DE devices. We have also demonstrated the production of an electrically conductive layer of carbonaceous combustion products by laser-scribing silicone rubber with a low-cost diode laser. Laser scribing allows for arbitrary electrode patterns with high detail and requires no masks or stencils. The technique can be integrated into the 3D printing process to produce electrodes on layers of printed elastomer. Unfortunately, silicone elastomer was found to be unsuitable for this process as the silicon compounds formed during combustion of the elastomer are extremely brittle and lose interconnectivity and thus electrical conductivity even under very small strain. Future investigation will focus on finding more suitable elastomer materials that can be both 3D printed and laser-scribed to form more compliant electrodes. If successful, this will result in a fully automated manufacturing process for DE devices that uses no molds or part-specific tools and requires no organic solvents, on a very simple, low-cost 3D printing system that can be built for under $1000 from readily available components.

4. EXPERIMENTAL SECTION

4.1 Materials

Silopren UV Electro 225-1 UV-curable silicone rubber, kindly supplied by Momentive Performance Materials Inc. (Waterford, NY, USA) was used as insulating/dielectric base material in 3D printing. Momentive UV LSR CAT is used as catalyst for UV curing. UV Electro 225-1 is made specifically for high-voltage applications. It has a Shore A hardness of 25, elongation at break of 580%, dielectric constant of 3.13 and dielectric breakdown strength of 24 kV/mm (unstretched). The uncured material has a viscosity of 70 Pa·s at 20°C. It can be fully UV-cured in seconds or minutes, depending on irradiation intensity. The pot life of UV Electro 225-1, once catalyst has been added, is 24 h at 20°C according to the manufacturer, however, many samples were found to still be liquid and usable after more than a week. Long pot life and short curing time are very useful properties for 3D printing.

Carbon conductive grease (MG Chemicals, Surrey, BC, Canada) was used as purchased in dual-material test prints.

GO aqueous dispersion (5.16 mg/mL) was kindly supplied by Rico Tabor (Monash University, Clayton, Australia).

GO/silicone composites were prepared by freeze-drying GO aqueous dispersion, re-dispersing dry GO in tetrahydrofuran (THF) under ultrasonication in an ultrasonic bath for 24 h, and then mixing with UV Electro 225/1 base diluted with THF. The mixture was sonicated for 1 h and then heated to 50°C for 24 h to evaporate solvents. Finally, UV LSR CAT was added at a ratio of 100:2, stirred and degassed in a vacuum chamber for 10 min before use.

To prepare GNP composites, expanded graphite purchased from Graphitene Ltd. (Stevenage, UK) was dispersed in chloroform at 50 mg/mL and sonicated in an ultrasonic bath for 24 hours. The dispersion was then centrifuged for 1 minute at 3000 rpm to sediment larger particles. The supernatant was decanted and then added in appropriate proportions to UV Electro 225/1 base diluted with chloroform. The mixture was sonicated for 1 h and then heated to 50°C for 24 h to evaporate solvents. Finally, UV LSR CAT was added at a ratio of 100:2, stirred and degassed in a vacuum chamber for 10 min before use.

4.2 3D printer

The 3D printer is custom-built and based on the open-source Prusa i3 design. The metal frame and other mechanical parts were purchased from Orballo Printing (Vigo, Spain). A Duet Wifi controller board and Duex5 extension board (Think3DPrint3D Ltd, Peterborough, UK) are used to drive the motors and UV light. The printer has a maximum print volume of 110 x 130 x 120 mm. The extruders use disposable auger valves, purchased from Adhesive Dispensing Ltd. (Milton Keynes, UK), to control material flow rate. Each extruder is driven independently by a stepper motor. Materials are fed into the valves from 10 mL UV-block syringes mounted on the print head. Any liquid or paste-like material with a viscosity between 10 Pa·s and 500 Pa·s can be dispensed. Tapered nozzles with 0.41 mm diameter were used to deposit layers between 100 µm and 300 µm thickness. To ensure continuous material flow, the syringes were pressurized to 100 mbar for UV Electro 225-1 and 300 mbar for carbon grease. A LED Engin LZ4-04UV00 UV LED (LED Engin, Inc., San Jose, CA, USA) with peak wavelength between 365 nm and 370 nm and a maximum radiant flux of 3.8 W is used for UV curing. Cure time depends on the size of the part. A layer is cured by scanning over the area with the UV LED at 5 mm distance above the surface of the deposited material. A 200 µm layer of UV-curable silicone rubber (see below) is fully cured after an exposure time of 5 s/cm².

4.3 Laser scribing

For laser scribing, a commercial Benbox mini laser engraver is used. The engraver is equipped with a 250 mW, 405 nm diode laser and driven by two micro stepper motors. It has a work area of 70 mm by 70 mm, scan speed up to 1000 mm/min and PWM control to adjust laser power. The focus of the laser is adjusted manually. Laser scribing of silicone rubber was performed at maximum power (continuous mode). The heat induced by the laser was regulated via the speed of the laser head. Benbox software was used to control the engraver.

4.4 Resistance measurements

Bulk conductivity of silicone composites was measured using a Keithley 2400 SourceMeter (Keithley Instruments Ltd., Bracknell, UK) and a four-point probe setup. Sheet resistance of laser-scribed films was measured using a Fluke 177 digital multimeter (Fluke Corporation, Everett, USA). Two flat copper electrodes were placed across a 2-mm-wide strip of laser-scribed film with a 2 mm gap between them to leave a square patch of film between the electrodes. In this setup, the measured resistance between the electrodes corresponds to the sheet resistance of the film (subject to influence from contact resistance). Resistance of the bending sample was measured using the Keithley 2400 SourceMeter in 2-wire sensing mode because this has a higher measurement range up to 211 MΩ.

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